

The coefficients of mass transfer accompanying absorption of carbon dioxide gas by distilled water in the slug flow regime are measured.

The slug flow regime in pipes occurs in many industrial situations. Many investigators have studied the hydrodynamic characteristics of such flow regimes. These results are on the whole generalized in [1-4]. Much less attention has been devoted to processes on the interphase boundary, in particular, interphase mass transfer. We should mention studies of mass transfer from a single slug [5, 6], as well as studies devoted to the mass transfer for slug flow in a gas lift [7, 8] and in a slug flow in a vertical pipe [9]. Thus, the absorption of a mixture of carbon dioxide gas and nitrogen by water in pipes 4.8 and 2.4 mm in diameter and 0.5-1 m long in the gas lift regime with short slugs was studied in [7]. It should be noted that in [7] it was erroneously concluded that the contribution of the mass from the lower part of the slug, where waves are observed on the interphase boundary, to the total flow is small. Thus, it was shown experimentally in [10] that waves on the surface of liquid films intensify mass transfer. The mass transfer coefficients were underestimated apparently because in the case of low velocities of the slug and liquid in [7] the flow around the slug is continuous, and behind the slug, as shown in [6], there exists a thin, distinct diffusion wake. Therefore mass transfer from the next slug occurs under conditions of a lower concentration head. The absorption of hexane in a mixture with different inert gases by kerosene in vertical pipes 6 and 8 mm in diameter and from 5 to 50 cm long was studied experimentally in [9]. The gas slugs were short. It was found that the mass transfer is strongly affected by the coalescence of slugs and the presence of an initial section of formation of the flow regime.

The results of the studies performed do not give the required information for carrying out sufficiently reliable calculations of mass transfer under conditions of slug flows in pipes. Therefore, the purpose of this work was to measure experimentally the coefficients of mass transfer in a wide range of values of the flow parameters and to determine the specific effect of flow parameters such as the length of the charge, the velocity of the liquid phase, and the frequency of the slugs on mass transfer.

Absorption of carbon dioxide gas by distilled water was used to model the mass transfer process. The carbon dioxide gas is weakly soluble and therefore the mass transfer is determined by the diffusion in the liquid phase. All hydrodynamic parameters of the regime were measured simultaneously in the experiments: the flow rate of the liquid and gas at the inlet, the temperature of the liquid, the velocity, length, and repetition frequency of the slugs. The concentration of the carbon dioxide gas dissolved in the water at the inlet and outlet of the working section was recorded continuously with conductometric sensors. Special experiments were also performed on measuring the thickness of the film of liquid between the gas slug and the pipe wall for different fluid velocities.

The experimental setup, a diagram of which is shown in Fig. 1, consists of the following basic parts: lower level tank, pump, mixer 1, gas slug former 3, working part 2, sample extractor 7, upper level tank, illuminator 4, diaphragms 5, section for recording the length and velocity of the slugs 6, and meters for measuring the flow rates of the liquid and gas.

The liquid consisted of distilled water, whose temperature was stabilized and equalled $20 \pm 0.2^\circ\text{C}$. The liquid was pumped into a mixer and then into the working part. The flow rate was measured with rotameters.

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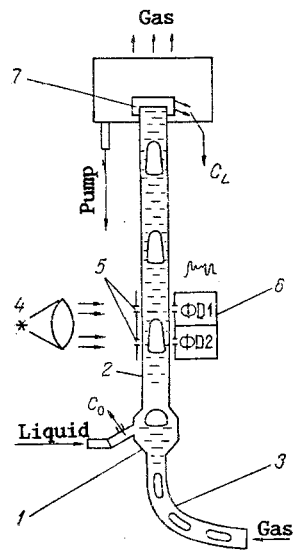


Fig. 1. Diagram of experimental setup.

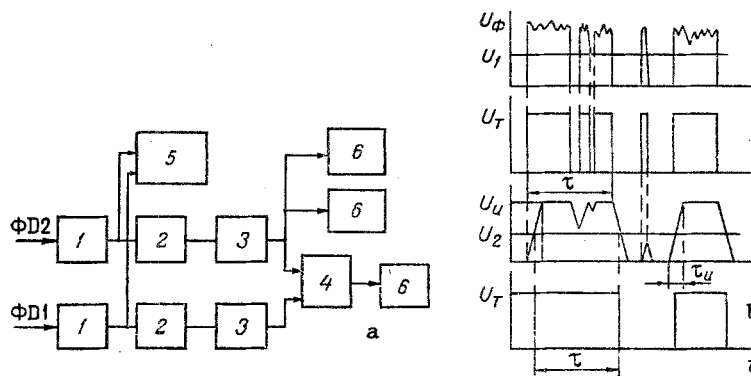


Fig. 2. Block diagram of the meter measuring the velocity, length, and frequency of slugs (a) and voltage diagrams of the operation of the block diagram (b).

A difficult problem in carrying out the model tests in the slug regime is the creation of conditions at the inlet such that the slugs would have the same length. For this, the gas was injected from a cylinder through the flow meter and flow-rate regulator into the slug former, and then into the mixer. The slug former consisted of a transparent flexible tube, lying almost horizontally and connected to the mixer. The tube was filled with the working liquid. The gas, passing along the horizontal part of the former, is collected into bubbles of equal volume. By changing the angle of inclination and the diameter of the tubes of the slug former the length of the slugs can be regulated over a wide range. Histograms of the slug lengths show that for moderate flow rates of the liquid the deviation of the slug length from the mean length is small. Thus, in regimes up to $Re = 2000$ and $\beta = 0.3$ more than 80% of the slugs fall within $\pm 5\%$ of the mean. For $Re = 3000$ the spread increases up to $\pm 15\%$.

The mixer consists of a section of pipe 30 mm in diameter with smooth transitions at the inlet and outlet and side connections for injecting the liquid. The gas bubbles were fragmented in mixers of a different type, and it was not possible to obtain regimes with identical slugs.

The working section consisted of a glass tube 1.5 m long and an inner diameter of 12.1 mm. The inner diameter was measured by the volume method. At the outlet from the working section all of the liquid was poured into the sample extractor, from which part of the solution was injected continuously into a conductivity gauge in order to measure the concentration of the dissolved carbon dioxide gas.

The velocity, frequency, and length of the slugs were determined by the stroboscopic method. Two photodiodes, positioned at some distance along the flow, were illuminated by

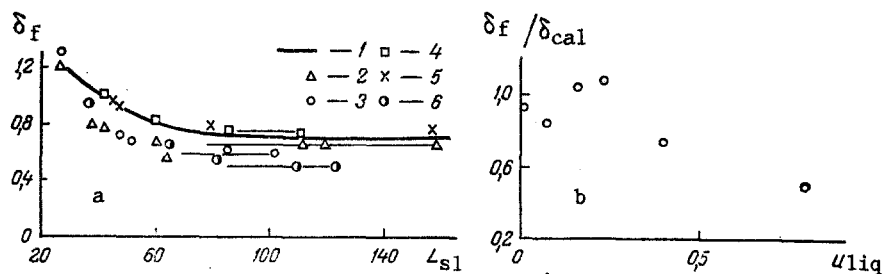


Fig. 3. Film thickness versus the slug length and fluid velocity. (a) 1) Calculation based on [12]; 2) $u_{liq} = 0$; 3) $u_{liq} = 8$ cm/sec; 4) 16; 5) 24; 5) 40 cm/sec; (b) δ_f / δ_{cal} versus u_{liq} for the developed section of the film. δ , mm; L_{sl} , mm; u_{liq} , m/sec.

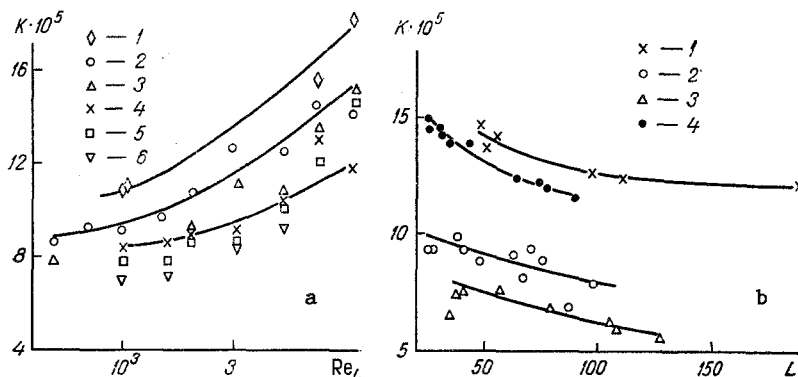


Fig. 4. The mass transfer coefficient versus Re and the slug length: (a) 1) $\beta = 0.2$; 2) 0.3; 3) 0.5; 4) 0.6; 5) 0.7; 6) 0.8; (b) 1) $Re = 3 \cdot 10^3$; $\beta = 0.3$; 2) 500 and 0.3; 3) 500 and 0.5; 4) single slug [4]. K , m/sec; L , mm.

a light source 4 (Fig. 1) through two horizontal slit diaphragms 5, a glass working tube with the working solution, and two point diaphragms. The distance between the diaphragms varied depending on the velocity and required measurement accuracy. The real signal from the photodiode has a spread associated with the presence of liquid bridges in the gas slug and bubbles in the liquid samples, so that the signals after the photodiodes were fed into a two-channel pulse forming circuit (Fig. 2a). It consisted of a Schmidt trigger 1, an integrator 2, a second Schmidt trigger 3, an analog circuit for forming pulses proportional to the velocity of the slugs 4, a loop oscillograph 5, and frequency meters 6. Each channel of the pulse former operates in the following manner. The first Schmidt trigger of the circuit is triggered by a voltage U_1 (Fig. 2b). The threshold voltage U_1 is regulated manually with the help of an oscillograph based on the smallest number of overshoots. The signal is then fed into the integrator. The integration time is regulated and chosen so that spikes from the bridges and bubbles would not intersect the average line U_2 . The voltage U_2 equalled one-half the saturation voltage U_u , and this voltage activated the second trigger. Thus the duration of the pulses from the second trigger equals the duration of the starting pulse. The outputs of both channels were connected to the analog circuit for forming pulses whose duration equalled the transit time of the slugs between the diaphragms. With the help of the frequency meters measurable pulses from 10 or 100 slugs can be averaged. The operation of the circuit was checked by the stroboscopic method - photographing of the moving slug with a double flash. It turned out that, as the number of bubbles and liquid bridges increases, when the flow rates increase the measurement error increases. This is attributable to the appearance of overshoots on the flat sections of the integrator pulses. In addition, the circuit can give interruptions, since unfiltered pulses from the bridges and bubbles switch the analog circuit at moments which do not correspond to the real boundaries of the slug. In this case, the signals after the first triggers were recorded on the loop oscillograph, and all of the information was obtained from the oscillograms.

The concentration of the dissolved carbon dioxide gas at the inlet and outlet of the working section was measured by the conductivity method [10]. The direct conductometric method is based on a single-valued dependence of the conductivity of the solution on the concentration of the solute. For each specific gauge a calibration curve of the conductivity versus the concentration, based on which the concentration of the solute was determined in the experiments, was constructed. The conductivity gauges consisted of a platinum wire 0.5 mm in diameter, welded into the glass so that a free end 5 to 7 mm long, which functioned as the sensitive element, remained. The gauge was inserted into a stainless steel housing, through which the working solution was continuously pumped. The gauges were calibrated by chemical titration of a 0.01 N carbon dioxide solution by a solution of sodium hydroxide with phenolphthalein as the indicator. The calibration was monitored before the start of each experiment.

The mass transfer coefficient was calculated from the logarithmic-mean difference of the concentrations [11]:

$$K = \frac{Q}{S_n} \ln \frac{C_s - C_0}{C_s - C_L}$$

In calculating the area of the surface it was assumed that the slug is shaped like a cylinder with a spherical head and a flat bottom. The diameter of the slug equals $d - \delta$. The thickness of the film was measured for an air slug rising in the moving liquid. For this, a portion of air with known volume was fed into the mixing setup with a known fluid velocity. The recording apparatus fixed the length and velocity of a single slug. By changing the volume of the slug and subtracting small values from high values, it is possible to calculate the average thickness of the film of the remaining part of the gas slug. For the slug with minimum size the tip was assumed to be hemispherical:

$$\delta = \frac{d}{2} - \frac{1}{2} \sqrt{\frac{4}{\pi} \frac{V_2 - V_1}{L_2 - L_1}}$$

where V_2 , V_1 and L_2 , L_1 are, respectively, the volumes and lengths of the gas slugs. The velocity of the liquid varied from 0 to 0.8 m/sec. The solid line in Fig. 3a shows the computed dependence from [12]. For all fluid velocities the initial section did not exceed five tube diameters. Figure 3b shows the relative thickness of the film for the cylindrical part of the slug.

The method used for measuring the slug velocity made it possible to measure the relative rise velocity of the slug u_∞ both in the stationary and moving liquid. The deviation from the well-known dependence $u_\infty = 0.345\sqrt{gd}$ [3] equals 10%. It should also be noted that the velocity u_∞ for carbon dioxide gas is about 5% lower than for air. This effect was pointed out in [13].

The mass transfer coefficients were measured in a wide range of values of the flow parameters of the slug flow. Reynolds number, constructed from the reduced velocity of the liquid, varied from 500 to 10^4 , and the volume gas content in the flow varied from 20 to 80%. Analysis of the data presented in Fig. 4a shows that for all gas concentrations the mass transfer coefficient increases monotonically as the velocity of the carrying phase increases. There is a substantial separation of the experimental points as a function of the gas content. In this series of experiments the length of the slugs was established in an arbitrary manner, and the spread in the slug lengths exceeded 100%. Each experimental point is an average over a large number of slugs of different length. In order to determine the effect of parameters such as the slug length, fluid velocity, and repetition frequency of the slugs on the mass transfer an experimental model of developed slug flow with a constant slug length and constant slug repetition frequency was realized. The number of bubbles in the liquid bridges was minimum, even at significant velocities. The length and frequency of the slugs could be regulated with fixed flow rates of the liquid and gas. Figure 4b shows the data on the effect of the slug length on mass transfer for different velocities and gas concentrations. For comparison, the experimental dependence for mass transfer from a single slug for a tube with the same diameter from [6] is shown. In all regimes the coefficient of mass transfer depends strongly on the slug length. As the velocity decreases this dependence becomes weaker. The mass transfer coefficient decreases as the gas concentration increases and it increases as the fluid velocity increases. The effect of the velocity is so strong that the mass transfer coefficients are higher than the coefficients for a single slug.

The process of mass transfer from the slugs into the moving liquid can be represented as follows. Each slug, within its residence time in the measurement section of length L , will leave a wake with dissolved gas of length

$$l = u_{\infty} \cdot \frac{L}{u_g(1 - \varphi)}$$

Increasing the velocity and reducing the gas content decrease the number of slugs present in the wake of preceding slugs. In the limit a single slug regime is realized. In addition, as the fluid velocity increases the fluid velocity at the interface with a constant relative velocity of the phases increases, and the velocity profile in the liquid film forms at larger distances than for a slug rising in a stationary liquid. This follows, for example, from an analysis of oscillograms of the instantaneous value of the friction at the wall for slug flow in a pipe 15 mm in diameter, presented in [4]. The direction of flow changes at a distance of approximately 50 mm, and the friction becomes negative. In turbulent flow regimes the effect of velocity pulsations is apparently negligible, since the pulsations decay rapidly when the flow, moving past the slug, rapidly narrows, which can also be clearly seen on the oscillograms presented in [4].

Thus the interphase mass transfer in slug flow depends on the length, frequency, and velocity of the slugs. All these quantities are interrelated with one another, and the degree to which the mass transfer is affected is determined by the specific conditions of the flow regime. The effect of the degree of mixing of the concentration in the wake requires further study.

NOTATION

β , delivered volume gas content; Re , Reynolds number; U_1 , U_2 , and U_u , voltages; N , normality of the solution; K , mass transfer coefficient; Q , volume flow rate of liquid; S_n , surface area of n slugs; C_s , equilibrium concentration on the surface of the liquid; C_0 , starting concentration; C_L , average concentration at the outlet from the pipe; d , inner diameter of the pipe; δ , thickness of the film; V , volume of the slug; L , length of the slug; u_{∞} , rise velocity of the slug in a stationary liquid; u_g , velocity of the slug; and ϕ , true volume gas content.

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